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FIRST QUARTERLY REPORT

Reporting Period 21 May 1963
to 19 August 1963

DEVELOPMENT OF HIGH ENERGY DENSITY PRIMARY BATTERIES
200 Watt Hours Per Pound
of Total Battery Weight
Minimum

Contract Number: NAS3-2775

LIVINGSTON ELECTRONIC CORPORATION

Montgomeryville, Pennsylvania

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by

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to

NATIONAL AERONAUTICS AND SPACE ADMINISTRATION
Lewis Research Center
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October 28, 1963

Mr. John J. Fackler, Contracting Officer
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Space Power Systems Procurement Section
Lewis Research Center
21000 Brookpark Road
Cleveland, Ohio, 44135

RE: Contract No. NAS3-2775

Dear Mr. Fackler:

We are enclosing the First Quarterly Report revised and delivered in accordance with instructions received from your staff.

Yours very truly,

C. E. Smiley
President
Livingston Electronic Corporation

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INTRODUCTION

This first quarterly progress report describes the research being conducted under Contract NAS3-2775 on the development of high energy density primary batteries.

The objective of this research program is to develop a battery of 200 watt hours per pound of total battery weight, which we are taking as a minimum. In the initial stages of the program, a selected study of recorded information was undertaken by our investigators to seek out nonaqueous systems which are likely to yield high watt-hours per pound.

Due to the broad scope of this entire assignment, a basic re-evaluation of thinking with respect to battery design is being attempted from a theoretical viewpoint.

As a result of the literature study, the theoretical studies, and initial experimental work, three significant new areas of exploration have been uncovered:

(1) An equation of state estimating the energy density capability of solvents in terms of three conveniently measured parameters has been evolved.

(2) The availability of ion exchange membranes permits practical consideration of alkaline anolytes and acid catholytes within the same cell. More complex electrolyte structures should provide for the utilization of lower weight and higher voltage electrodes.

(3) A new approach to the formation of conducting solutions in solvents of lower dielectric constant has been formulated.

PART I

QUANTITATIVE DESCRIPTION OF PROGRESS

The goal of this research program is to develop a battery of 200 watt hours per pound of total battery weight. The theoretical deliberations and literature studies constituted the bulk of the effort during this initial period. The results of these efforts appear interesting and in several instances somewhat unexpected.

The dielectric constant of the primary solvent will probably not have to be high provided that the coordination requirements of the conducting solute are met. This permits the consideration of such battery electrolytes as common hydrocarbons containing an alkali halide coordinated with an atmosphere of a highly polarized compound. If the secondary solvation sheath of the ions still requires the action of a high dipole material, a small quantity of such normally solid substances as acetamide may be dissolved into the primary solvent whereupon its migration to the surfaces of the charged species provides an activity far out of proportion to the actual concentration of the secondary ligand.[1]

It is therefore planned to measure conductivity in many primary solvents irrespective of the dielectric constant value but in the presence of unusual atmospheres such as NH_3, SO_2, and other polar materials substantially below their vapor pressures. When favorable conductivities have been obtained, the effects of solid polar additives may be expected to provide further conductivity increases.

Conductivity at the expense of electrolyte decomposition is not desirable. While such a solute as LiF does not display the best solubility characteristics, it may be generally expected to resist electrolytic decomposition. While NH_3, SO_2, and other polar ligands as liquids may decompose easily in many circumstances, their characteristics when firmly coordinated to a charged ion may be totally different.[2]

In the theoretical considerations, an attempt was made to resolve the overall battery problem into a single equation of state with respect to the properties of the solvent. To a large measure, this has been accomplished yielding a figure of merit, watt hours per pound, from literature or experimental values of specific conductivity and modified Tafel constants characterizing electrolytic decomposition.

The uniform collection of conductivity and decomposition data for numerous solvents at varying ligand pressures has commenced. These data will be evaluated by the equation of state to provide figures of merit for each combination, using five solutes ranging from lithium fluoride to tetra-butyl-ammonium chloride. Currently, the data is being collected manually. Automatic equipment is planned for increased data production rate. See Schematic 1.

Three practical ammonia test cell vehicles have been previously developed by Livingston Electronic Corporation at the Montgomeryville Laboratory. Since these vehicles are suited for the exclusion of air and moisture and operation above one atmosphere, they are suitable for use with a wide variety of solvents. Testing of complete cells has been initiated in these vehicles in anticipation of a major effort in this direction during latter periods. Ion exchange membranes were tested in liquid ammonia cells providing increased energy efficiency.

1. Abstract Control #4004
2. Ind. & Eng. Chem., 55, 9 p. 15-28 (Sept., 1963)

PART II

LITERATURE STUDY

Activities in this phase of the task have resulted in collection, analysis, abstracting, and coding of the pertinent and useful information on batteries and electrochemical systems.

In this survey, considerable emphasis has been placed on solvent systems in that the electrolyte must be compatible with highly active, light weight electrodes for use in high energy density batteries.

The staff has developed a system of information retrieval in the fields of nonaqueous batteries, separator materials, special cells, materials of construction, statistical, thermodynamic, and engineering evaluations to determine favorable areas for high energy density battery performance, nonaqueous electrolytic studies, nonaqueous theoretical electrochemistry, batteries in general, and electrolytic studies in general.

The literature search is not intended to be all-inclusive, but rather selective as to pertinent material. It will encompass summaries, bibliographies, and reviews in areas of interest. It concentrates on chemical abstracts, reports from the Armed Services Technical Information Agency (ASTIA), OTS, PIC, and other related information agencies. The selection has been governed by considerations of proper worth as well as applicability. The limits and depth of selection are controlled by definitions and terms relating to battery construction and performance properties in nonaqueous and aqueous systems.

Basic operations undertaken in order to specifically document the battery literature include acquiring graphic records of potential interest, abstract bulletins, patents, government and commercial reports, technical trade publications, journals, and reviews dealing primarily with electrochemistry and batteries.

The staff has covered much of the above area by utilizing the library facilities of:

(a) Livingston Electronic Corporation

(b) G. & W. H. Corson Company, Inc.

(c) Philadelphia Free Library System (Main Branch)

(d) The Franklin Institute Library

The libraries listed above subscribe to important indexing and abstracting services in the principal area of interest. The following sources for information have been included in the search:

1. Chemical Abstracts (C.A.)
2. Power Information Center (P.I.C.)
3. Office of Technical Services (O.T.S.)
4. Armed Services Technical Information Agency (ASTIA)
5. Department of Commerce Bulletins
6. Extended Abstracts from Battery Division of the Electrochemical Society
7. Applied Science and Technology Index
8. Journal of Physical Chemistry
9. Journal of Electrochemical Society
10. Chemical & Engineering News
11. Physics Abstracts
12. Metallurgical Abstracts (London)
13. Doctoral Abstracts in Chemistry, Physics, and Engineering
14. Engineering Abstracts
15. Periodic Guide to Literature
16. Battelle Abstracts
17. Physical Reviews
18. Foreign Translations of Communist Block Countries (O.T.S.)

A method for the analysis of the acquired records as the basis for organizing them in such a way as to simplify the identification of wanted information has been established by generally evaluating:

(1) The aspect of the record (article, patent, book) which is likely to be of interest to persons engaged in battery research.

(2) The method in which these aspects should be expressed in the retrieval system in order to match the way potential users are apt to think and express their requests for information.

A bibliography of literature reviews is given in Reference I.

PART III

MATERIAL COMPATIBILITY EVALUATION

A total of 314 short-term tests has been completed to screen the effects of 19 selected solvents on 27 separator materials.

In most cases, the materials have been placed in a test tube, the solvent added and the tube stoppered with glass wool and set aside for a sufficient period of time to yield the desired information. See Plate I. For volatile solvents, an aluminum foil vapor barrier was placed at the mouth of the tubes. To test materials at room temperature in liquid ammonia and other low boiling solvents, it was necessary to store the tubes in pressure chambers.

A summary of the results of the completed tests is given in Table I.

Compatibility of the materials with p-fluorotoluene, Freon 114, aromatics, and a silicone oil were good to excellent. Most of the separators appeared compatible with ammonia except two ion exchange membranes. Cyclohexanone, propylene carbonate, and pyridine were hard on ion exchange materials but SA 6404, a strong acid membrane, held up in all solvents to which it was exposed. Cellulosic, polyalkane, and epoxy fibre papers gave generally good performance. Butylamine, nitromethane, cyclohexanone, butyrolactone, and n-methyl-pyrrolidone were increasingly severe.

Plate I shows the equipment used for material compatibility evaluations with fixed solvents. The work with solvents which boil substantially below room temperature indicates that low temperature tests are inadequate. Many materials appear stable at low temperature and subsequently fail at room temperature where it is necessary to apply pressure to retain the liquid solvent but yet provide for periodic examination. Techniques have been evolved for this work and data are included for ammonia and Freon 114 exposures. Essentially, the method consists of storing the materials for test inside the test tubes charged with the liquid solvent at low temperature.

These tubes are sealed in the stainless steel quick coupler chambers (See Plate II), pressurized to immobilize the solvent, and allowed to stand at room temperature. Examination is affected by refrigeration and venting of pressure. Re-exposure is then practical.

PART IV

A GENERAL ENERGY CONSIDERATION OF THE ROLE OF THE SOLVENT

A theoretical approach is being evolved to indicate the maximum energy density that might be obtained from the many solvent system combinations under consideration. This approach utilizes basic data for the solvent in an "equation of state" to estimate an upper limit for the watt hours per pound available from the majority of cells which could be constructed using a particular solvent system. The factors electrode potential, solvent decomposition, conductivity, drain rate, discharge time, etc., are generalized in terms of a single independent variable for treatment by the calculus.

Though this theoretical method is presently incomplete and subject to further revision, it indicates that a generalized approach concerning the solvent is feasible and of practical value in designing electrolytes.

The definitions and symbols used are listed at the end of the following derivation which is based upon one pound of electrode reactants.

$$(\text{output}) = (\text{cell voltage}) * (\text{cell current}) * (\text{time})$$

$$(\text{cell voltage}) = (\text{cell potential}) - (\text{resistive losses})$$

$$\text{Let } X = (\text{cell potential})$$

$$\text{and } (\text{resistive losses}) = IRL/A$$

$$(\text{cell voltage}) = (X - IRL/A)$$

$$(\text{cell current}) = (\text{theoretical current}) - (\text{corrosion current})$$

$$\text{Let } I = \text{theoretical current}$$

$$(\text{corrosion current}) = JA$$

where J = corrosion current density and

A = electrode surface area

$$(\text{cell current}) = (I - JA)$$

Let T = time in consistent units and

W = output, in watt hours

Therefore:

$$W = (X - IRL/A)(I - JA)(T)$$

Expansion and rejection of the last term gives:

$$W = XIT - XJAT - I^2RTL/A$$

Let $IT \doteq Fe(HX - S)$

where $e(HX - S)$ means 2.72 to the $(HX - S)$ power.

$H \doteq 0.4$ and $S \doteq 5.5$ a limiting empirical relationship for the electrodes.

Let $J \doteq e\left(\frac{X-C}{D}\right)$

Using the functional notation as above with C being the Tafel constant plus free energy and ionization contribution to the decomposition voltage. The symbol D is the Tafel slope.

Inclusion of the above terms gives:

$$W = XFe(HE - S) - XTAe\left(\frac{E-C}{D}\right) - \frac{F^2RL}{AT}\, e(2HE - 2S)$$

Where F = Faraday's constant in ampere hours/ pound

This equation as it stands may be solved for the maximum value of W by iteration through numerical values of X and T. The application of calculus is expected to reduce this to an iteration in X only or perhaps to a direct literal solution. The conductivity and decomposition data being collected will be fitted into the final form of this equation selected to screen out the electrolyte combinations showing sufficient promise as to qualify them for evaluation in experimental cells and high energy density primary batteries.

LIST OF SYMBOLS USED

X = Cell potential

I = Theoretical current (Stoichiometric)

J = Corrosion current density

A = Electrode surface area

JA = Corrosion current

T = Time in consistent units

W = Output in watt hours

L = Inter-electrode spacing

R = Electrolyte specific resistance

F = Faraday's constant in ampere hours/pound

S & H = constants See Appendix I

C & D are modified Tafel constants measured for each electrolyte.

PART V

CONDUCTIVITY AND DECOMPOSITION POTENTIAL MEASUREMENTS

An experimental study of promising electrolyte systems, with special emphasis on nonaqueous solvents, complex salts, and coordination compounds has been initiated. Currently, this study includes the following measurements:

(1) The electrical conductivity of the electrolyte (solvent and solute).

(2) The decomposition potential or breakdown voltage of the electrolyte.

The equipment used for measuring the conductivity of these "special solutes" - nonaqueous solvent combinations consists of: special stainless steel chambers, nylon conductivity cells, teflon conductivity cells, bright platinum electrodes, constant voltage line transformer, vacuum tube voltmeter, and a variable frequency audio generator. See Plate III.

Lithium fluoride may be the most desirable salt from the decomposition voltage aspect, but its solubility is generally poor. Approximately 6 volts will be required to decompose this salt; hence, such decomposition as may be observed could be attributed to the solvent. The following salts were selected as tools for a uniform searching of many solvents:

lithium chloride	lithium fluoride
potassium bromide	potassium fluoride
potassium iodide	magnesium bromide
tetramethyl ammonium chloride	aluminum chloride hydroxylamine hydrochloride

The decomposition potential study has been initiated both with the solvents and with several solute-solvent combinations. Table II presents the decomposition values available at this time.

The techniques and equipment for measurement of conductivity and decomposition potentials for a broad spectrum of

solvents irrespective of vapor pressures have been evolved and the first decomposition data was available in time for inclusion in this report. This data refers to saturated solutions and is uncorrected for electrode surface area and resistive losses which will subsequently be subtracted from the applied voltage. Consequently, no interpretations are presented at this writing.

PART VI

INCLUSION OF ION EXCHANGE MEMBRANES IN LIQUID AMMONIA CELL

Experimental evidence has shown that the inclusion of specific ion exchange membranes within the electrode stack has yielded significant improvements in the cell discharge characteristics when compared with all other separator materials used in existing liquid ammonia systems.

Separator Background - In the past separators of cellulose, Teflon, Nylon, Dynel, glass, polyethylene, polypropylene, and many other organic and inorganic compounds have been utilized. The selection of a membrane was determined not only by the compatibility of the separator in the solvent system, but by pore size and thickness. High-rate batteries required membranes of large pore sizes and thin cross sections. Membranes of the materials listed would not prevent corrosion nor anode attack on long active stand nor would they prevent lead or mercury bridging from the cathode to the anode. However, the high-rate batteries developed in the past had short life requirements and accordingly, their limited discharge life was acceptable.

Low rate long life batteries utilized these same materials for separation but they were required to be of greater cross section thickness, and smaller pore size in order to prevent excessive self-discharge, reduce corrosion, and diminish accumulation of nonconducting films.

Batteries of intermediate rate required separator materials which showed a compromise between the two extremes above described.

Separator Design - The problem of separator choice becomes very important, especially when very active anode/cathode couples are contemplated. The Mg anode in liquid ammonia is rapidly attacked by an acid present in the cell. Acid ion is often required by the cathode for high coulombic efficiencies.

For example, in the Mg/KSCN:NH_3/S:C/Ag cell the sulfur cathode in the presence of the NH_4^+ ion is approximately two times as efficient as when no NH_4^+ or other acid ion is included. The paradox of anode corrosion in the presence of the acid and increased cathode efficiency in the presence of the acid makes the selection and design of separator materials very difficult. The corrosion problem has been, in the past, overcome partially by the use of massive anodes. As the large Mg anode is dissolved in the ammonia solution, 6 moles of ammonia ($Mg \cdot 6NH_3$) are combined with the Mg, thus consuming large quantities of solvent. The large anode and excess solvent required by this cell or battery severely diminish the high theoretical energy available from this system. The inclusion of the ion exchange membrane in the cells has reduced the anode corrosion greatly so that smaller anodes and less solvent are now required in the cell.

Identification of Component Parts, Steps, Explanation of Mode of Operation of System, and Test Results - The use of an ion exchange membrane adjacent to the anode in liquid ammonia cells has shown greatly improved discharge characteristics. The Gelman SA 6404 cation exchange membrane adjacent to the anode in the Mg/$HgSO_4$ cell has shown an increase in watt hours per pound of net cell as follows:

Control Cell Mg/$HgSO_4$ at -70°F. 18.05 WH/lb. in 4.5 hours

Ion Exchange Cell Mg/$HgSO_4$ at -70°F. 52.7 WH/lb. in 8.4 hours

The unique feature of the functioning of these membranes in liquid ammonia permits a considerable reduction in cell volume, also, as indicated by the following data:

Control Cell Mg/$HgSO_4$ at -70°F. 1.22 WH/in.^{3}

Ion Exchange Cell Mg/$HgSO_4$ at -70°F. 2.36 WH/in.^{3}

The inclusion of an ion exchange membrane increased the cathode coulombic efficiency from 42.8% to 75.6%. This exemplifies the

improved values; no effort has been made to optimize the weight of Mg anode and other materials of construction. A significant improvement in energy density will be realized when parameters of material combinations are optimized including minimizing of weight.

The mercuric sulfate depolarized cell has a low theoretical energy density. For this reason, the same ion exchange membrane cell technique will be transferred to the Mg/KSCN/S:C/Ag system where the sulfur has an extremely high theoretical energy density (excess of 500 WH/lb.). This may be regarded as indicating the feasibility of achieving 70-80% efficiencies with the sulfur cathode thus yielding significantly higher energy densities. Cell discharge curves comparing both ion exchange cells and control cells are shown in Figures 1 and 2. Table III is shown for comparison of: discharge time, voltage, current density, weight, coulombs achieved, efficiency, and watt-hours per pound. The use of the anion exchange membrane, which rejects the positive Mg^{++} ion is shown in Figure 1. It is to be noted that all of this data is reported at -70°F. Coulombic efficiencies and voltage regulation at room temperature and at +120°F. is usually identical with the data shown at -70°F.

Construction of both control cells and ion exchange membrane cells is shown in Plate Numbers IV and V.

Initial tests utilizing the cation exchange membranes in the Mg/KSCN/S:C/Ag system have yielded promising results but optimization of S:C salt ratios has not been completed at this time. The initial indications at this time are encouraging in that a significant increase in coulombic efficiencies was achieved.

Control Cell Mg/S	7.84% coulombic efficiency
Ion Exchange Cell Mg/S	17.0% coulombic efficiency
Control Cell Mg/S	26.2 watt-hours per pound
a. Ion Exchange Cell Mg/S	58.4 watt-hours per pound
b. Ion Exchange Cell Mg/S	70.0 watt-hours per pound

It must be kept in mind that these data are taken at -70°F. and that the cell components are not optimized as to weights, mix ratios, or spacing.

PART VII

SPECIAL APPROACHES

As the theoretical and literature studies proceeded, specific areas have emerged which exhibit a reasonable probability for the development of high energy density batteries. In the early period of the study it was expected that these approaches would constitute exceptions to the general battery considerations. For example, Appendix I describes an empirical relationship between equivalent weight and electrode potential. Sulfur is the most favorable exception to this generalization, particularly with respect to a solvent such as ammonia, which exhibits a strong resistance to reduction. Unfortunately, sulfur chemistry can be exceedingly complex and a surprisingly large portion of the available theory on sulfur chemistry is debatable. Early emphasis has been placed in this work on the application of the sulfur cathode in ammonia test cells. Since control of the difficult S / NH_3 chemistry will be paramount, this work commenced with the introduction of ion exchange membranes into the better understood Mg/KSCN:NH_3/$HgSO_4$ cell as described in Part VI. Cells of the type Mg/NH_3/S have a tangible probability of achieving 200 watt hours per pound provided the complex S/NH_3 chemistry can be controlled.

One of the combinations being considered by Livingston Electronic Corporation for the development of a high energy density primary battery system is a lithium anode of a concentrated lithium ammonia bronze solution. In addition to lithium, the other alkali metals (sodium, potassium, rubidium, and cesium) and the alkaline earth metals (calcium, barium, and strontium) are soluble in liquid ammonia, carbon, methylamine, and fused salts. A quantitative investigation of the adsorption spectra of these solutions has shown that the spectra are identical at the same concentration regardless of the

alkali or alkaline earth metal used. The concentrated solutions are deep bronze in color and conduct an electric current as well as the free metal. At all concentrations, the equivalent conductance is greater than that found for any known solvent. Likewise, the conductance for concentrated solutions is extremely high and of the same order of magnitude as that found for metals; for example, the specific conductance of a saturated solution of sodium in liquid ammonia is 0.5047×10^{4} (ohm cm.)$^{-1}$ and the specific conductance of metallic sodium is 2.16×10^{5} (ohm cm.)$^{-1}$ at 20°C. and for mercury the specific conductance is 1.036×10^{4} (ohm cm.)$^{-1}$. The equivalent conductance goes through a minimum and then increases with dilution to a limiting value. The behavior of the equivalent conductance at higher dilutions is much the same as found for ordinary inorganic compounds in liquid ammonia or water.

Numerous investigators have studied the nature of electrical conductance in solutions of liquid ammonia-alkali and liquid ammonia-alkaline earth metals. It appears that most of the current is carried by "free electrons" and "solvated electrons." The reactions may be represented by:

$$\text{I.A} \quad Li + yNH_3 \longrightarrow Li(NH_3)_y^+ + e^-$$

$$\text{I.B} \quad e^- + zNH_3 \longrightarrow e(NH_3)_z^-$$

$$\text{II.} \quad M + yNH_3 \longrightarrow M(NH_3)_y^+ + e(NH_3)^-_{y-z}$$

Summarizing the results of all of the investigations, it appears that metal atoms, solvated metal ions, free electrons, and solvated electrons are present in equilibrium in these solutions.

In support of these conclusions, it is believed that the investigations of Farkas, Pauli, and Block are of particular value. A brief resume of their work is presented in Appendix II of this report. Other references pertinent to these concepts are listed in the literature survey. (See Code Entry 2, Card Column 127-137.)

It may be concluded from these investigations that at low temperatures and high concentrations, all of the lowest states in which the electrons can exist are filled and in these states the electrons are paired. Only the relatively small number in the higher states are unpaired and able to contribute to the magnetic susceptability; that is, orient themselves in a magnetic field. At high temperatures or low concentrations, respectively, more electrons can be in the higher states, or there are not enough electrons altogether to fill all possible states. Pauli-Block theory does not give quantitative agreement with the observed susceptability in the case of the alkali metals, but is does explain the essential features of the phenomena, namely, that the paramagnetism is small and nearly independent of the temperature.

Based on the above listed conclusions, there is a clear value in approaching the problem of a high energy power supply by the use of alkali and alkaline earth metal solutions in the anode compartment of the cell.

The kinetic advantages of using alkali or alkaline earth metals - liquid ammonia solution in the anode compartment of the cell stem from the inherent nature of the free electrons in solution and those that are partially cavitated by ammonia molecules. In the case where the electron is free, its energy is immediately available for electrochemical reactions. Its rate of discharge at the electrode should be independent of double-layer effects, electrode overpotential, and current density. For the solvated electrons, a certain amount of energy is necessary to break up the $e(NH_3)^-_{y-z}$ complex before the electron can become available for electrode reactions. The potential of the electrode depends directly on the behavior of the solvated electron and the properties of the transfer reaction. Many influences on the electrode potential of the electrode take place by an indirect change in the transfer reaction.

Retardation of the electron release from $e(NH_3)^-_{y-z}$ leads to a special kind of overpotential, the transfer overpotential. Retardation resulting from reaction overvoltage and diffusion overvoltage may be neglected for metal-liquid ammonia solutions.

The utilization of the alkali or alkaline earth metal solutions as anodes in high energy density batteries will require conducting electrolytes or ion exchange membranes which can resist the strong reducing action of the free electron. Ammonia has been discussed as an example of the first category. The glass pH electrode with its alkaline metal ion error at high pH values offers a second since glass is known to resist electrical attack as well. Thus cells of the type:

Li solution/ion exchange membrane/liquid Cl_2 or Br_2

will be given careful consideration since extremely high energies should be available if the proper materials and methods of construction can be found.

The above consideration is extended to cover next the hybrid solvent cells. Water as a solvent permits the action of strong acids but is limited with respect to bases by the basicity of the OH^- ion. Ammonia, on the other hand, allows extreme basicity but strongly levels acidity. The coupling of an ammonia anode to an aqueous cathode may provide considerable merit. An example which will be studied is:

$$Mg/NH_3:Salt/NH_3:H_2O:Salt/H_2O:Salt/MDNB$$

Thus it may be practical to combine the best halves of two different battery systems if the junctions can be adequately constructed.

Additional specialized approaches have been mentioned in previous reports and will be covered in greater detail as experimental data either supports or rejects the radical concepts involved.

PART VIII

WORK TO BE PERFORMED DURING THE NEXT PERIOD

1. During the Next Quarter

The literature search will continue at a decreasing rate. Group and individual study of the data will receive substantial effort. Methods of reducing the theoretical products to experimental cells will receive increased consideration. The generalized approach to the role of the solvent will be brought to a logical conclusion and used to screen the specific resistivity-decomposition voltage data for electrolytes favorable to high energy density battery requirements. Consideration of electrodes for favorable electrolytes will start from the thermodynamic viewpoint and attempts will also be made to formulate a generalized concept.

Experimental measurement of the conductivity and decomposition voltage parameters of a uniform set of solutes in numerous solvents and varying ligand atmospheres will be converted to automatic scanning and an increased volume of data is anticipated.

Testing of nonaqueous cells in the Livingston ammonia test cell vehicles will be given increased attention. Magnesium and lithium anodes will be considered in normal battery structures and in unconventional designs.

The materials evaluation program will continue with emphasis shifting from the general to specific needs and newly available materials. Efforts to obtain ion exchange materials for evaluation purposes will continue and foreign sources will also be considered. Additional solvent types will be added and classes showing little variation within the group will be restricted to one representative item. For example, aromatics, freons, and alkanes will be restricted to one compound in each group.

2. During the Next Month

The literature and theoretical solvent studies will continue. An attempt will be made to simplify the form and usage of the solvent oriented battery equation of state by functional notation and to obtain a numerically operational form.

Hardware and test equipment shown to be useful in the work to date will be set up in larger quantities to provide for increased experimental activity.

Application of ion exchange membranes to control S-NH_3 chemistry in Mg/NH_3/S cells will continue in an effort to gain higher efficiency from this theoretically high energy system.

The materials evaluation program will fill the majority of the gaps in Table I and evaluate the recently acquired Whatman ion exchange membranes.

PLATE I

MATERIAL COMPATIBILITY TEST

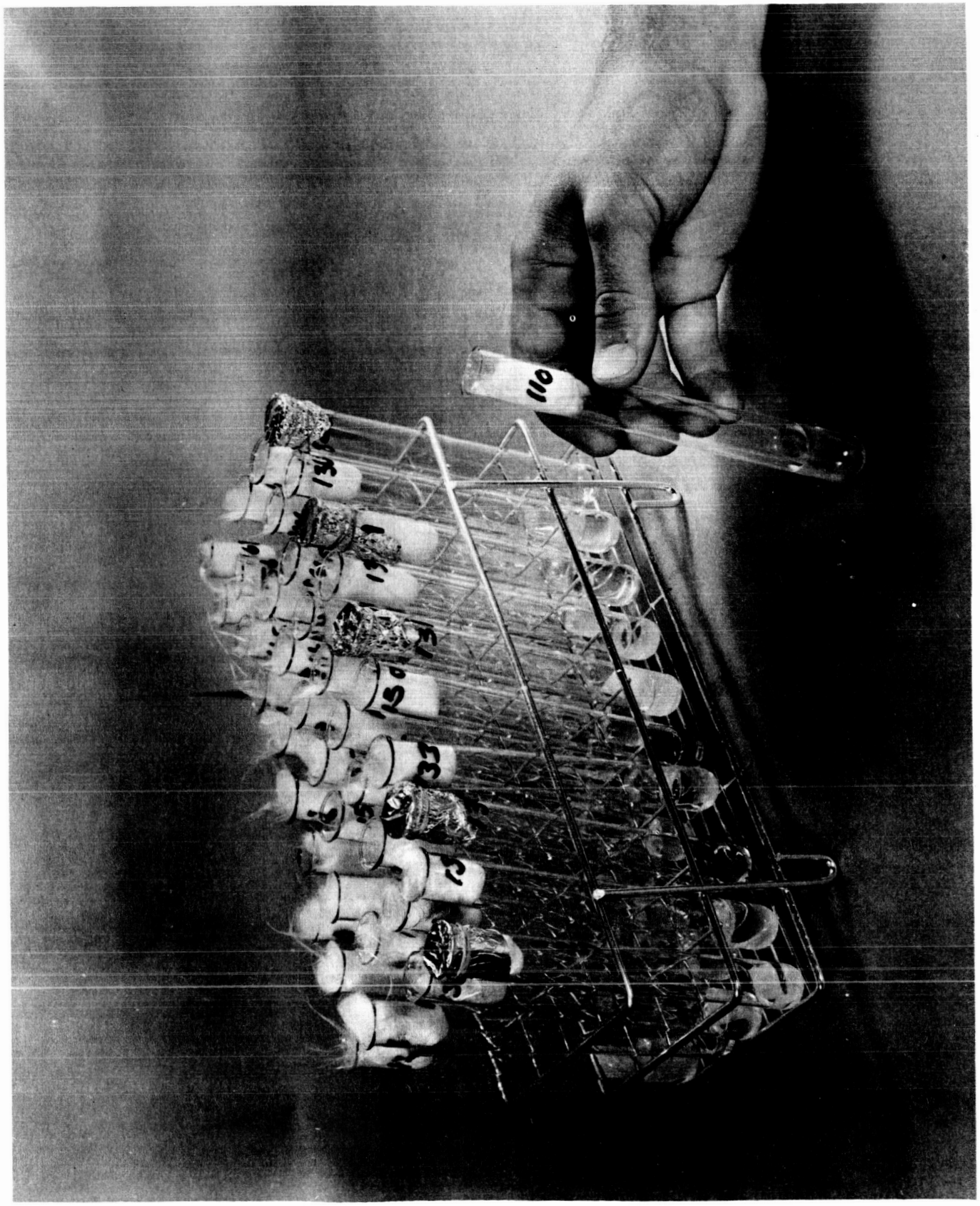


PLATE II

STAINLESS STEEL CHAMBER

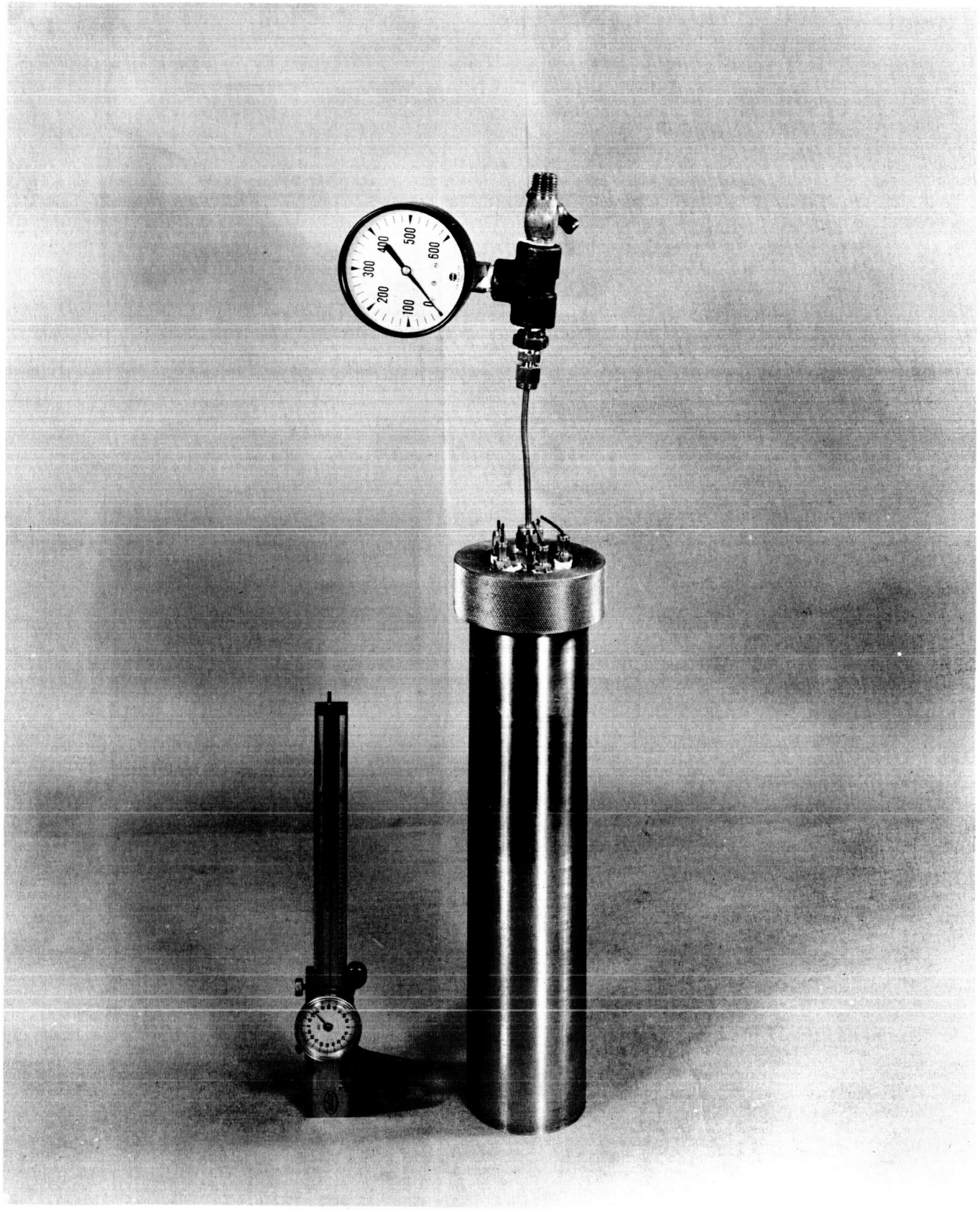


PLATE III

EQUIPMENT FOR CONDUCTIVITY MEASUREMENTS

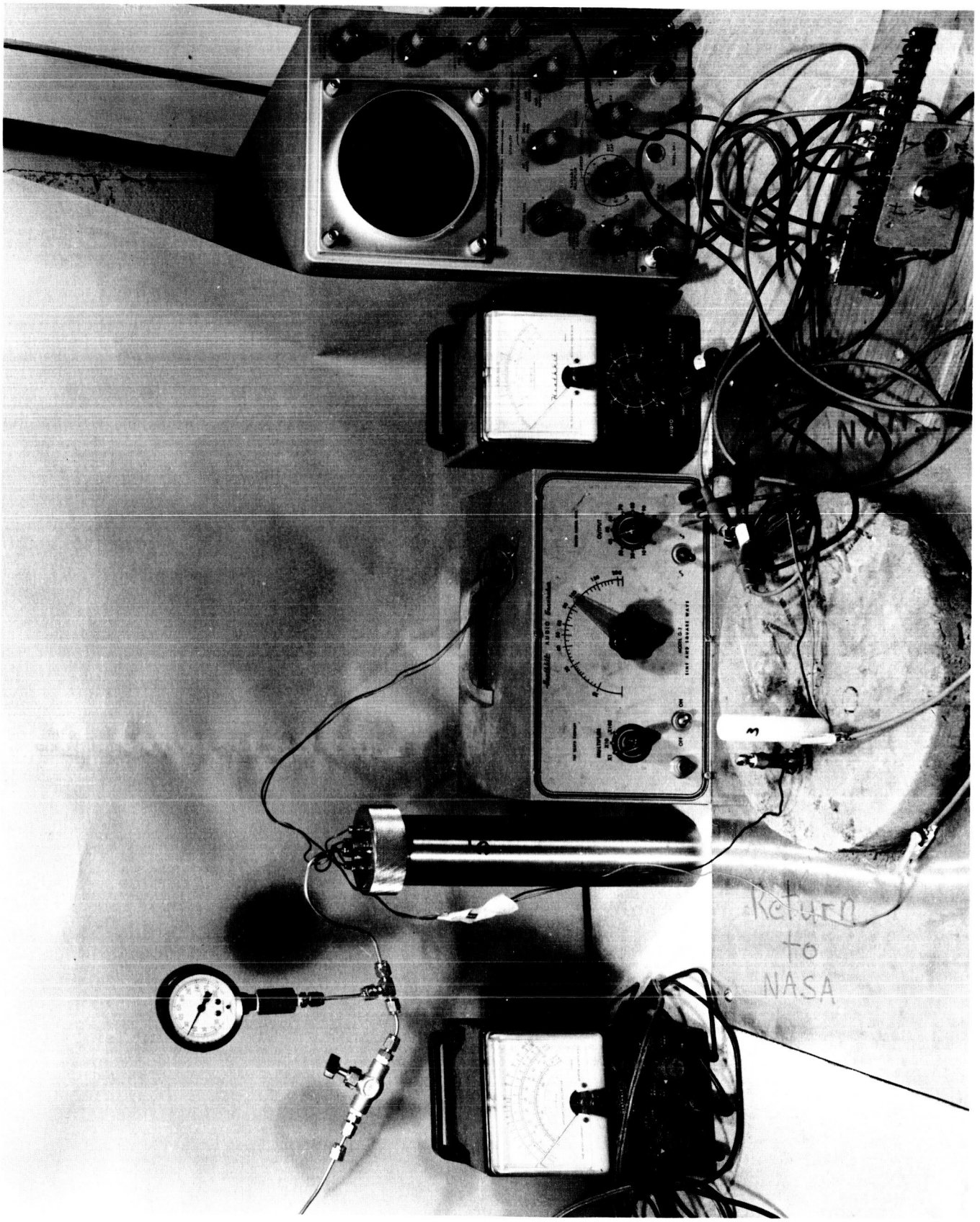


PLATE IV

CELLS

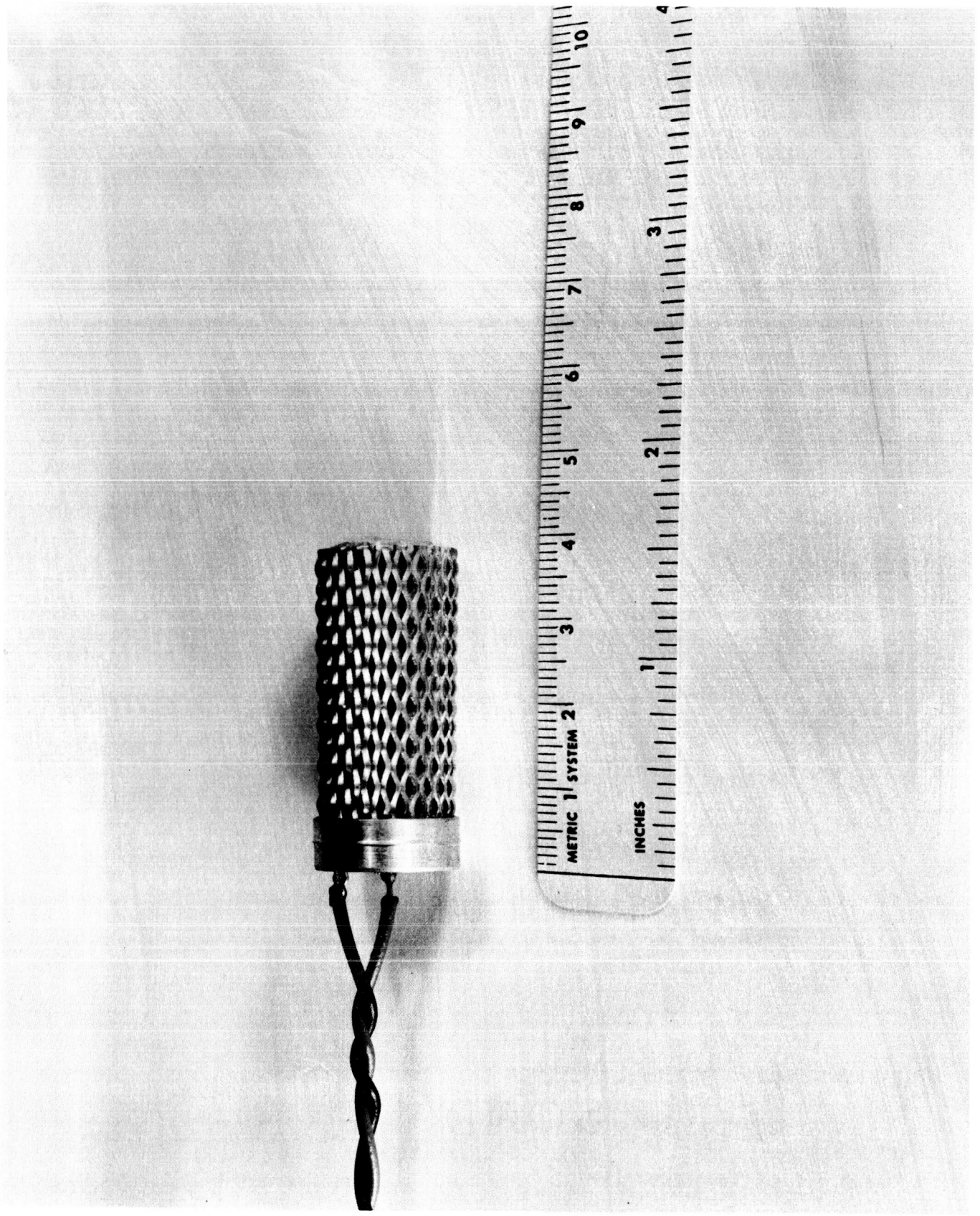
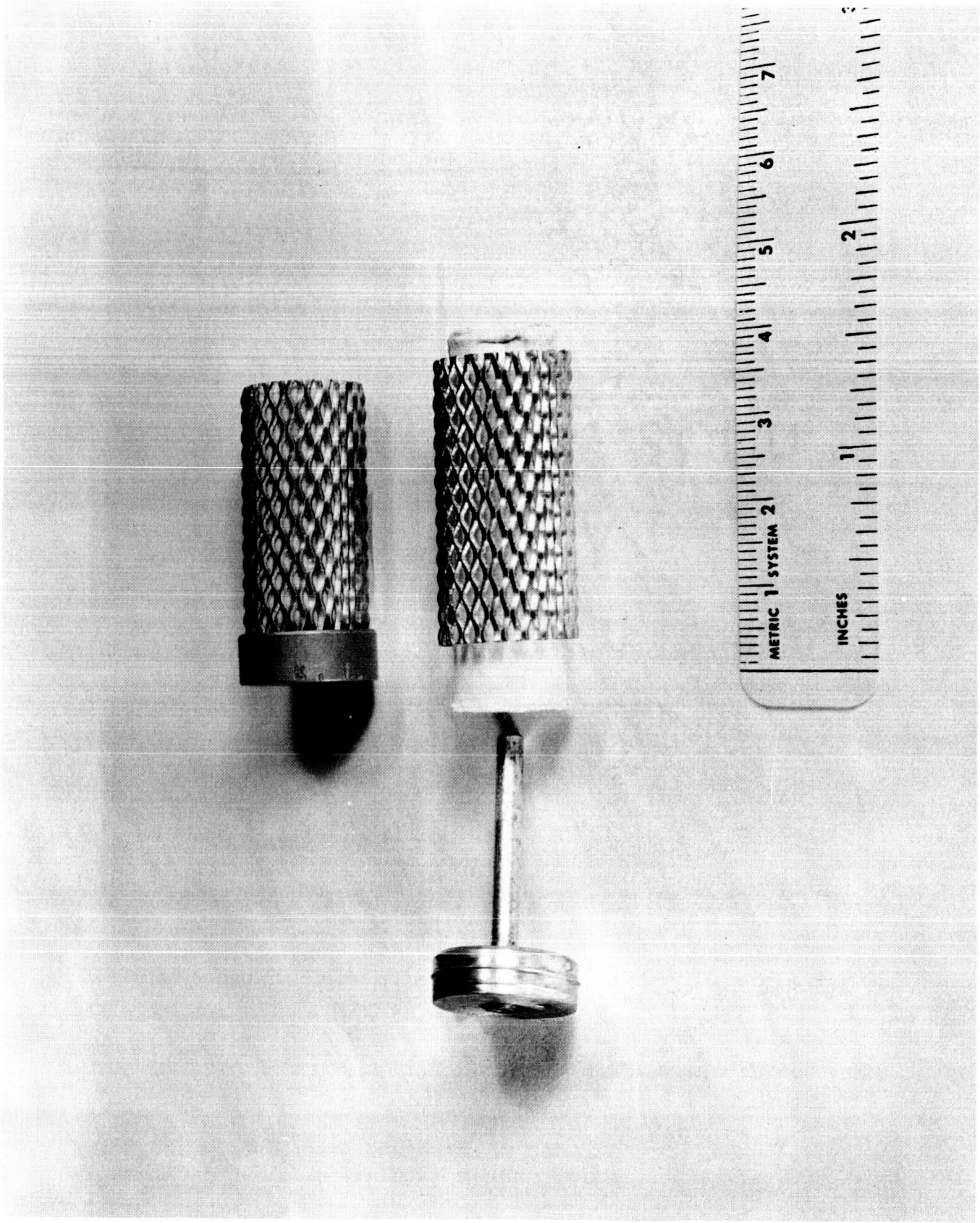


PLATE V

CELLS



IDENTIFICATION OF ION-EXCHANGERS AND SEPARATORS

Gelman Instrument Co., Ion-Exchange Membranes, "ACROPOR"

SB 6407 - On nylon
WA 6402 - On nylon
WB 6403 - On nylon
SA 6404 - On nylon
WA 6406 - On nylon

Ionac Chemical Co., Ion-Exchange Membranes

MC 3142 - Cation Membrane
MA 3148 - Anion Membrane
XLMC 3235 - Cation Membrane
XLMA 3236 - Anion Membrane

Rohm & Haas Co.

LA-1 - Liquid Ion-Exchanger - Anion - "Amberlite"
LA-2 - Liquid Ion-Exchanger - Anion - "Amberlite"

SEPARATORS

A. H. Thomas Co., Filter Papers

Whatman No. 42
Whatman No. 50

E. I. Du Pont De Nemours & Co.

Textryl - Calendered - Dacron — Production has been discontinued.
Textryl - Calendered - Dacron

Webril Non-Woven Fabrics, Kendall Co., Walpole, Mass.

EM 470 - Dynel — (Type M - Battery Grades)
M 1401 - Dynel
M 1410 - Dynel
M 1450 - Dynel
M 1406 - Nylon - Dynel Binder
EM 312 - Nylon - Dynel Binder
M 1216 - Viscose - Vinyl Binder
M 1231 - Viscose - Chandler Binder

TABLE NO. 1

MATERIALS

Column No. (See Code & Comments)	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21	22	23
Solvents			Butyrolactone	n-Methyl-2-Pyrrolidone	Liquid Ammonia	Propylene Carbonate	Isopropylamine	p-Fluorotoluene	Freon 11	Freon 113	Freon 114	Pyridine	Benzene	Toluene	Xylene	Propylamine	Butylamine	Nitromethane	Cyclohexanone	DC-200 Silicone Oil	P-Fluorotoluene		
Days Under Test			18	18	5	4		5			4	5	4	5	5		5	5	5	4	5		
6407	IE	n	R	R		R						R	E	E	G		E	R	R				
6402	IE	n	R	R	G	R					G	R	E	E	E		E	R	R				
6403	IE	n	R	R		R						R	E	E	E		E	R	R				
6404	IE	n	G	G	G	G					G	G	E	E	E		E	G	G				
6406	IE	v	R	R	G	R					G	R	E	E	E		E	R	R				
3142	IE				R						G												
3148	IE				R						G												
3235	IE				G						G												
3236	IE				G						G												
Whatman 42	S	1	E	E		E						E	E	E	E		E	E	E				
Whatman 50	S	1	E	E								G	E	E	E		Fb	E	G	E			
Textryl-C	S	d	G	G		G						Fs	E	E	E		Fb	E	Fs	E			
Textryl-F	S	d	G	Fs		G																	
EM470	S	a	D	D								D	G	E	E		E	D	D	E			
M1401	S	a	D	D		U		E															
M1410	S	a	D	D																			
M1450	S	a	D	D																			
M1406	S	n	Fb	Fb								Fb	G	Fs	E		G	Fb	Fs	Fs			
EM312	S	n	Fb	Fb		Fb		G				Fb	G	G	E		Fb	Fs	Fs	E			
M1216	S	v	Fm	Fb	G	G						G	G	G	G		U	G	G	E			
M1231	S	v	Fs	Fm		G						G	G	E	E		Fb	E	E	E			
EM478	S	v	Fm	Fm		G						U	Fs	G	E		U	Sw	Fm				
M1365	S	c	Fb	Fb	Sw	Fs		G				G	E	E	G		G	G	E	E			
EM476	S	pp	E	E	G	G						G	Fs	E	G		Sw	Sw	Sw				
R2205	S	l			Sw	G		G				E	E	E	E		E	E	E	E			
(Bags)	S	pe			E	G						Fs	E	E	E		U	E	E				
Ultipor 9	S	i				G						G	E	E	E		Fs	E	E				
Epocel 10	S	i				G						Fs	Fm	Fs	Fs		U	Fs	Sw				
IPC1478	S	i				Fs																	

CODE

Column 1
IE - Ion Exchanger
S - Separator

Column 2
n - Nylon
d - Dacron
v - Viscose
a - Dynel
c - Cotton
l - Cellulose
i - Impregnated Paper
pp - Polypropylene
pe - Polyethylene

Columns 3 to 21
E - Excellent
G - Good
M - Moderately Good
D - Dissolved
B - Binder Dissolved
Fs - Frayed or Disintegrated Slightly
Fm - Frayed or Disintegrated Moderately
Fb - Frayed or Disintegrated Badly
R - Ion-Exchanger Partly Removed from Fabric
Sw - Swollen
U - Unsatisfactory

COMMENTS

Col. #3: #6407-Trace of yellow in solution
6403-Yellow p'p't
Textryls-Bright yellow solutions
EM470-Slight yellow solution
M1401-Bright yellow solution
M1410-Bright yellow solution
M1450-Bright yellow solution
EM476-Slight yellow solution

Col. #4: 6403-Slight yellow p'p't
M1410-Trace of undissolved fibers

Col. #5: M1365-Not disintegrated
R2205-Good

Col. #6: M1401-Gelatinous and weak

Col. #12: M1365-Gelled

Col. #13: All good after 1 hour

Col. #17: M1231-Soft pulp
M1365-Soft pulp
IPC1478-Soft pulp

Col. #19: IPC1478-Probably good

EM 478 - Viscose Gauze - Chandler Binder
M 1365 - Cotton - Chandler Binder - Balance Acetate
EM 476 - Polypropylene
R 2801 - Pure Cellulose
R 2901 - Pure Cellulose
R 2401 - Pure Cellulose
R 2205 - Pure Cellulose
Polypropylene Sheet - 0.020" Thick
Polyethylene Sheet - From Bag Stock

Pall Corporation, Epoxy Resin Impregnated Papers

Ultipor 9
Epocel - 3 micron
Epocel - 10 micron
Epocel - 30 micron

Knowlton Brothers, Paper:

IPC 1478 - Impregnated with Dibutoxyethyphthalate

Whatman Ion Exchange Membranes:

Manufactured by W. & R. Balston, Ltd., England

Notes

1 -	Anion Exchanger - Ecteola Cellulose	Paper ET20
2 -	Anion Exchanger - Aminoethylcellulose	Paper AE30
3 -	Anion Exchanger - Diethylaminoethylcellulose	Paper DE20
4 -	Cation Exchanger - Cellulose Phosphate	Paper P20
5 -	Cation Exchanger - Carboxymethylcellulose	Paper CM50

1 - Registered in U. S. Patent Office
2 - Registered in U. S. Patent Office
3 - Registered in U. S. Patent Office
4 - British Patent No. 838973
5 - Registered in U S. Patent Office

COMMENTS RELATIVE TO TABLE I

Material Compatibility Evaluation

At the end of the test period, the ion exchange and separator materials were examined while in the solvents and after having been removed therefrom and dried.

A summary of the test results completed to date is given in Table No. 1. From this this table, certain general conclusions have been drawn about the completed tests.

1. All of the materials that were tested in p-fluorotoluene, Freon 114, benzene, toluene, xylene and DC-200 silicone oil were found to be compatible.

2. All of the materials which were tested in liquid ammonia, except #3142 and #3148 ion exchangers, were satisfactory.

3. In propylene carbonate, most ion exchangers, a dynel, and one nylon fabric were unsatisfactory. Two-thirds of the materials were good.

4. Pyridene had an adverse effect on most of the ion exchangers, dynel, nylon, one viscose, and cotton. Ten materials were satisfactory.

5. In Butylamine, all ion exchangers seemed to be unaffected. However, the viscose separators, the cotton and two of the impregnated papers failed.

6. Nitromethane dissolved dynel and partially destroyed three ion exchangers and one nylon. 72% of the materials survived the exposure.

7. Cyclohexanone, also, dissolved dynel and partially removed the resins from three ion exchangers. 78% of the materails were all right.

8. In butyrolactone and in n-methyl-2-pyrrolidone, only one ion exchanger, cellulose, viscose and polypropylene withstood the tests.

9. Of the ion exchangers, it should be noted that SA 6404 performed well in all solvents, and WA 6406 was second best.

10. The membranes that did not fail in any solvent are: Whatman filter paper, polyproylene, polyethylene, R-2205 cellulose and Epocel 10.

11. The materials that failed only in the butylamine are: two viscose fabrics, Ultipor 9 and IPC 1478.

(The manufacture of Textryls has been discontinued; so no further consideration shall be given to them.)

TABLE II

DECOMPOSITION VALUES

Solutes

E = Voltage
I = Current in Micro-amperes

Solvent	Solvent Only		Lithium Chloride		Tetramethyl Ammonium Chloride		Sodium Bromide		Potassium Bromide	
	E	I	E	I	E	I	E	I	E	I
N-Butylamine	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	0.5	0.0	0.5	0.5	0.5	0.0	0.5	0.0	0.5	0.0
	1.0	0.0	1.0	1.3	1.0	0.0	1.0	0.0	1.0	0.0
	1.5	0.0	1.5	1.8	1.5	0.0	1.5	0.0	1.5	0.0
	2.0	0.0	2.0	3.0	2.0	0.0	2.0	0.0	2.0	0.0
	2.5	0.3	2.5	3.8	2.5	0.0	2.5	0.0	2.5	0.0
	3.0	0.5	3.0	4.0	3.0	0.5	3.0	0.0	3.0	0.0
	3.5	0.5	3.5	4.3	3.5	0.5	3.5	0.0	3.5	0.0
	4.0	0.5	4.0	5.0	4.0	0.8	4.0	0.0	4.0	0.0
	4.5	0.5	4.5	6.0	4.5	0.8	4.5	0.0	4.5	0.0
	5.0	0.5	5.0	6.0	5.0	1.0	5.0	0.0	5.0	0.0
Pyridine	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	0.5	0.3	0.5	3.0	0.5	3.0	0.5	1.0	0.5	1.3
	1.0	2.0	1.0	11.0	1.0	12.0	1.0	2.0	1.0	2.2
	1.5	2.2	1.5	24.0	1.5	25.0	1.5	6.3	1.5	3.0
	2.0	3.0	2.0	35.0	2.0	46.0	2.0	16.0	2.0	4.3
	2.5	4.0	2.5	70.0	2.5	73.0	2.5	30.0	2.5	6.0
	3.0	5.0	3.0	> 100	3.0	> 100	3.0	48.0	3.0	7.0
	3.5	6.0					3.5	74.0	3.5	9.0
	4.0	7.0					4.0	> 100	4.0	10.8
	4.5	8.0							4.5	13.0
	5.0	9.0							5.0	15.0
Propylamine	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	0.5	0.0	0.5	4.0	0.5	0.0	0.5	0.0	0.5	0.0
	1.0	0.3	1.0	10.0	1.0	0.0	1.0	0.0	1.0	0.0
	1.5	0.5	1.5	18.0	1.5	0.0	1.5	0.0	1.5	0.0
	2.0	1.0	2.0	28.0	2.0	0.0	2.0	0.0	2.0	0.0
	2.5	1.75	2.5	40.5	2.5	0.5	2.5	0.0	2.5	0.7
	3.0	1.8	3.0	51.5	3.0	1.0	3.0	1.0	3.0	1.0
	3.5	2.0	3.5	68.0	3.5	1.0	3.5	1.0	3.5	1.0
	4.0	2.0	4.0	86.0	4.0	1.5	4.0	1.8	4.0	1.0
	4.5	2.0	2.0	> 100	4.5	1.8	4.5	2.0	4.5	1.0
	5.0	2.0			5.0	1.8	5.0	2.0	5.0	1.0

Solvent	Solvent Only		Lithium Chloride		Tetramethyl Ammonium Chloride		Sodium Bromide		Potassium Bromide	
	E	I	E	I	E	I	E	I	E	I
Iso-Propylamine	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	0.5	0.0	0.5	1.0	0.5	0.0	0.5	0.0	0.5	0.0
	1.0	0.0	1.0	2.0	1.0	0.0	1.0	0.0	1.0	0.0
	1.5	0.5	1.5	2.5	1.5	0.3	1.5	0.0	1.5	0.0
	2.0	0.8	2.0	3.5	2.0	1.8	2.0	0.0	2.0	0.3
	2.5	1.8	2.5	4.0	2.5	1.9	2.5	0.3	2.5	0.0
	3.0	1.8	3.0	4.5	3.0	2.0	3.0	0.3	3.0	0.0
	3.5	2.0	3.5	5.0	3.5	2.0	3.5	0.5	3.5	0.0
	4.0	2.0	4.0	6.0	4.0	2.0	4.0	0.5	4.0	0.0
	4.5	2.0	4.5	7.5	4.5	2.5	4.5	0.8	4.5	0.3
	5.0	2.0	5.0	8.0	5.0	3.0	5.0	1.0	5.0	1.0
Propylene Carbonate	0.0	0.5	0.0	0.0			0.0	0.0	0.0	0.0
	0.5	4.0	0.5	5.0			0.5	1.0	0.5	2.0
	1.0	13.0	1.0	22.0			1.0	2.0	1.0	4.0
	1.5	23.0	1.5	60.0			1.5	4.3	1.5	8.5
	2.0	44.0	2.0	>100			2.0	10.0	2.0	16.3
	2.5	73.0					2.5	17.0	2.5	28.0
	3.0	>100					3.0	26.0	3.0	40.0
							3.5	48.0	3.5	54.0
							4.0	81.0	4.0	68.3
							4.5	>100	4.5	86.0
									5.0	>100
Ethyl Acetate	0.0	0.0			0.0	0.0	0.0	0.0	0.0	0.0
	0.5	1.5			0.5	1.5	0.5	0.0	0.5	0.0
	1.0	3.0			1.0	2.5	1.0	0.0	1.0	0.0
	1.5	4.0			1.5	6.0	1.5	0.5	1.5	0.3
	2.0	5.0			2.0	8.0	2.0	0.7	2.0	1.0
	2.5	6.0			2.5	11.0	2.5	1.0	2.5	1.0
	3.0	8.0			3.0	14.0	3.0	1.3	3.0	1.8
	3.5	9.0			3.5	18.0	3.5	1.7	3.5	2.0
	4.0	10.0			4.0	22.0	4.0	2.0	4.0	2.0
	4.5	11.0			4.5	26.0	4.5	2.0	4.5	2.0
	5.0	13.0			5.0	30.0	5.0	2.0	5.0	2.0
N-Methyl 2-Pyrolidone	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	0.5	2.0	0.5	1.5	0.5	6.0	0.5	1.0	0.5	1.0
	1.0	3.0	1.0	5.0	1.0	17.0	1.0	3.0	1.0	4.0
	1.5	5.5	1.5	25.0	1.5	30.0	1.5	18.0	1.5	10.0
	2.0	7.5	2.0	66.0	2.0	48.0	2.0	48.0	2.0	26.0
	2.5	10.0	2.5	>100	2.5	66.0	2.5	>100	2.5	47.0
	3.0	13.0			3.0	88.0			3.0	76.0
	3.5	16.0			3.5	>100			3.5	>100
	4.0	17.0								
	4.5	21.0								
	5.0	26.0								

Solvent	Solvent Only		Lithium Chloride		Tetramethyl Ammonium Chloride		Sodium Bromide		Potassium Bromide	
	E	I	E	I	E	I	E	I	E	I
Butyro-lactone	0.0	0.5	0.0	0.0	0.0	1.0	0.0	0.0	0.0	0.0
	0.5	3.0	0.5	2.0	0.5	6.0	0.5	1.8	0.5	1.8
	1.0	10.0	1.0	9.0	1.0	13.0	1.0	4.0	1.0	4.0
	1.5	34.0	1.5	46.0	1.5	43.0	1.5	15.0	1.5	11.5
	2.0	59.0	2.0	>100	2.0	88.0	2.0	42.0	2.0	29.0
	2.5	>100			2.5	>100	2.5	69.0	2.5	47.5
							3.0	>100	3.0	69.5
									3.5	>100
Mercapto-propionic Acid	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	0.5	0.0	0.5	2.0	0.5	6.0	0.5	0.0	0.5	0.0
	1.0	1.5	1.0	4.0	1.0	25.0	1.0	0.2	1.0	0.2
	1.5	2.0	1.5	6.0	1.5	63.0	1.5	0.5	1.5	1.0
	2.0	2.5	2.0	7.5	2.0	>100	2.0	1.0	2.0	1.5
	2.5	3.0	2.5	9.5			2.5	1.8	2.5	1.8
	3.0	3.7	3.0	11.0			3.0	2.0	3.0	1.8
	3.5	4.0	3.5	13.0			3.5	2.0	3.5	2.0
	4.0	4.5	4.0	15.0			4.0	2.0	4.0	2.0
	4.5	5.0	4.5	17.0			4.5	2.0	4.5	2.0
	5.0	5.5	5.0	19.0			5.0	2.2	5.0	2.1
Nitro-methane	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	0.5	2.0	0.5	1.5	0.5	2.0	0.5	2.0	0.5	1.0
	1.0	4.0	1.0	3.0	1.0	4.0	1.0	4.8	1.0	2.0
	1.5	14.0	1.5	11.0	1.5	12.0	1.5	10.0	1.5	8.0
	2.0	33.0	2.0	29.0	2.0	59.5	2.0	16.0	2.0	24.0
	2.5	54.0	2.5	58.0	2.5	>100	2.5	22.0	2.5	51.0
	3.0	84.0	3.0	>100			3.0	30.8	3.0	79.0
	3.5	>100					3.5	39.0	3.5	>100
							4.0	45.0		
							4.5	52.0		
							5.0	59.5		
Dichloro-toluene	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	0.5	0.0	0.5	0.0	0.5	0.0	0.5	0.2	0.5	0.8
	1.0	0.0	1.0	0.0	1.0	0.0	1.0	0.5	1.0	2.0
	1.5	0.0	1.5	0.0	1.5	0.5	1.5	1.5	1.5	2.5
	2.0	0.5	2.0	0.0	2.0	0.5	2.0	1.8	2.0	4.0
	2.5	0.5	2.5	0.5	2.5	1.0	2.5	2.0	2.5	4.7
	3.0	1.0	3.0	1.0	3.0	1.5	3.0	2.1	3.0	5.5
	3.5	1.5	3.5	1.5	3.5	1.5	3.5	2.3	3.5	6.0
	4.0	2.0	4.0	2.0	4.0	1.5	4.0	3.0	4.0	6.5
	4.5	2.0	4.5	2.0	4.5	1.8	4.5	3.8	4.5	8.0
	5.0	2.0	5.0	2.1	5.0	2.0	5.0	4.0	5.0	8.0

Solvent	Solvent Only		Lithium Chloride		Tetramethyl Ammonium Chloride		Sodium Bromide		Potassium Bromide	
	E	I	E	I	E	I	E	I	E	I
Tri-Chloro-benzene	0.0	0.0	0.0	0.0	0.0	0.0				
	0.5	0.0	0.5	0.0	0.5	0.0				
	1.0	0.0	1.0	0.0	1.0	0.0				
	1.5	0.0	1.5	0.0	1.5	0.0				
	2.0	0.0	2.0	0.0	2.0	0.0				
	2.5	0.0	2.5	0.0	2.5	0.0				
	3.0	0.0	3.0	0.0	3.0	0.0				
	3.5	0.0	3.5	0.0	3.5	0.0				
	4.0	0.0	4.0	0.0	4.0	0.0				
	4.5	0.0	4.5	0.0	4.5	0.0				
	5.0	0.0	5.0	0.0	5.0	0.0				
Fluoro-toluene	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0		
	0.5	0.0	0.5	0.2	0.5	0.0	0.5	0.0		
	1.0	0.0	1.0	1.3	1.0	0.0	1.0	0.0		
	1.5	0.5	1.5	2.0	1.5	0.5	1.5	0.0		
	2.0	1.0	2.0	2.0	2.0	0.0	2.0	0.0		
	2.5	1.8	2.5	3.0	2.5	0.1	2.5	0.0		
	3.0	2.0	3.0	3.8	3.0	0.2	3.0	0.0		
	3.5	2.2	3.5	4.0	3.5	0.5	3.5	0.0		
	4.0	2.8	4.0	4.8	4.0	1.0	4.0	0.0		
	4.5	3.5	4.5	6.0	4.5	1.3	4.5	0.0		
	5.0	3.8	5.0	6.2	5.0	1.8	5.0	0.0		
Parachloro-benzo-trifluoride	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0		
	0.5	0.0	0.5	0.0	0.5	0.0	0.5	0.0		
	1.0	0.0	1.0	0.0	1.0	0.0	1.0	0.0		
	1.5	0.0	1.5	0.0	1.5	0.0	1.5	0.0		
	2.0	0.0	2.0	0.0	2.0	0.0	2.0	0.0		
	2.5	0.0	2.5	0.0	2.5	0.0	2.5	0.0		
	3.0	0.0	3.0	0.0	3.0	0.0	3.0	0.0		
	3.5	0.0	3.5	0.0	3.5	0.0	3.5	0.0		
	4.0	0.0	4.0	0.0	4.0	0.0	4.0	0.0		
	4.5	0.0	4.5	0.0	4.5	0.0	4.5	0.0		
	5.0	0.0	5.0	0.0	5.0	0.0	5.0	0.0		
Hexylene Glycol	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	0.5	0.0	0.5	0.0	0.5	1.0	0.5	0.0	0.5	0.0
	1.0	0.0	1.0	1.2	1.0	1.0	1.0	0.0	1.0	0.0
	1.5	0.0	1.5	2.3	1.5	2.0	1.5	0.0	1.5	0.0
	2.0	0.0	2.0	5.0	2.0	4.0	2.0	0.0	2.0	0.0
	2.5	0.0	2.5	8.0	2.5	6.5	2.5	0.0	2.5	0.2
	3.0	0.0	3.0	14.2	3.0	10.0	3.0	0.5	3.0	0.4
	3.5	0.0	3.5	20.0	3.5	14.0	3.5	0.8	3.5	0.6
	4.0	0.1	4.0	25.8	4.0	18.0	4.0	1.0	4.0	0.8
	4.5	0.3	4.5	32.3	4.5	22.5	4.5	1.0	4.5	1.0
	5.0	0.5	5.0	39.5	5.0	26.0	5.0	1.8	5.0	1.3

Solvent	Solvent Only	Lithium Chloride	Tetramethyl Ammonium Chloride	Sodium Bromide	Potassium Bromide
Trichloro-ethane	0.0 0.0	0.0 0.0	0.0 0.0	0.0 0.0	0.0 0.0
	0.5 0.0	0.5 0.0	0.0 0.0	0.5 0.0	0.5 0.0
	1.0 0.0	1.0 0.0	0.0 0.0	1.0 0.0	1.0 0.0
	1.5 0.0	1.5 0.0	0.0 0.0	1.5 0.0	1.5 0.0
	2.0 0.0	2.0 0.0	0.0 0.0	2.0 0.0	2.0 0.0
	2.5 0.0	2.5 0.0	0.0 0.0	2.5 0.0	2.5 0.0
	3.0 0.0	3.0 0.0	0.0 0.0	3.0 0.0	3.0 0.0
	3.5 0.0	3.5 0.0	0.0 0.0	3.5 0.0	3.5 0.0
	4.0 0.0	4.0 0.0	0.0 0.0	4.0 0.0	4.0 0.0
	4.5 0.0	4.5 0.0	0.0 0.0	4.5 0.0	4.5 0.0
	5.0 0.0	5.0 0.0	5.0 0.0	5.0 0.0	5.0 0.0
Fluoro-dinitro-benzene	0.0 0.0	0.0 0.0	0.0 0.0	0.0 0.0	0.0 0.0
	0.5 0.0	0.5 0.2	0.5 0.0	0.5 0.0	0.5 0.0
	1.0 0.0	1.0 1.3	1.0 0.0	1.0 0.0	1.0 0.0
	1.5 0.5	1.5 2.0	1.5 0.0	1.5 0.0	1.5 0.0
	2.0 1.0	2.0 2.5	2.0 0.0	2.0 0.0	2.0 0.0
	2.5 1.5	2.5 3.0	2.5 0.0	2.5 0.0	2.5 0.0
	3.0 2.0	3.0 3.8	3.0 0.2	3.0 0.0	3.0 0.0
	3.5 2.2	3.5 4.0	3.5 0.5	3.5 0.0	3.5 0.0
	4.0 2.8	4.0 4.8	4.0 1.0	4.0 0.0	4.0 0.0
	4.5 3.5	4.5 6.0	4.5 1.3	4.5 0.0	4.5 0.0
	5.0 3.8	5.0 6.2	5.0 1.8	5.0 0.0	5.0 0.0

	Lithium Fluoride		Potassium Fluoride		Aluminum Sulfate	Lithium Chloride and Aluminum Sulfate	Tetra Methyl Ammonium Chloride and Hydroxy Amine Hydrochloride
	E	I	E	I			
N-Butylamine	0.0	0.0	0.0	0.0			
	0.5	0.0	0.5	0.0			
	1.0	0.0	1.0	0.0			
	1.5	0.0	1.5	0.0			
	2.0	0.0	2.0	0.0			
	2.5	0.0	2.5	0.0			
	3.0	0.0	3.0	0.0			
	3.5	0.0	3.5	0.0			
	4.0	0.0	4.0	0.0			
	4.5	0.0	4.5	0.0			
	5.0	0.0	5.0	0.0			
Pyridine	0.0	0.0	0.0	0.0			
	0.5	1.0	0.5	1.0			
	1.0	1.8	1.0	1.3			
	1.5	2.5	1.5	2.0			
	2.0	4.2	2.0	3.0			
	2.5	7.0	2.5	4.2			
	3.0	9.0	3.0	6.0			
	3.5	12.0	3.5	7.0			
	4.0	15.0	4.0	9.0			
	4.5	18.0	4.5	11.0			
	5.0	20.0	5.0	13.0			
Propylamine	0.0	0.0	0.0	0.0			
	0.5	0.0	0.5	0.0			
	1.0	0.0	1.0	0.0			
	1.5	0.0	1.5	0.0			
	2.0	0.0	2.0	0.0			
	2.5	0.0	2.5	0.0			
	3.0	0.0	3.0	0.0			
	3.5	0.0	3.5	0.0			
	4.0	0.0	4.0	0.0			
	4.5	0.0	4.5	0.2			
	5.0	0.0	5.0	0.5			
Iso-Propylamine	0.0	0.0	0.0	0.0			
	0.5	0.0	0.5	0.0			
	1.0	0.0	1.0	0.0			
	1.5	0.0	1.5	0.0			
	2.0	0.0	2.0	0.0			
	2.5	0.0	2.5	0.0			
	3.0	0.0	3.0	0.0			
	3.5	0.0	3.5	0.0			
	4.0	0.0	4.0	0.0			
	4.5	0.0	4.5	0.0			
	5.0	0.0	5.0	0.0			

	Lithium Fluoride		Potassium Fluoride		Aluminum Sulfate		Lithium Chloride and Aluminum Sulfate		Tetra Methyl Ammonium Chloride and Hydroxy Amine Hydrochloride
	E	I	E	I	E	I	E	I	
Propylene Carbonate			0.0	0.2	0.0	0.0	0.0	1.0	
			0.5	2.0	0.5	5.0	0.5	7.0	
			1.0	4.0	1.0	12.0	1.0	23.0	
			1.5	8.0	1.5	24.0	1.5	62.0	
			2.0	12.0	2.0	46.0	2.0	> 100	
			2.5	17.5	2.5	73.0			
			3.0	22.0	3.0	> 100			
			3.5	28.0					
			4.0	34.0					
			4.5	40.0					
			5.0	45.0					
N-Methyl 2-Pyrolidone	0.0	0.0	0.0	0.0					
	0.5	1.5	0.5	1.0					
	1.0	3.0	1.0	2.0					
	1.5	4.0	1.5	5.0					
	2.0	6.0	2.0	8.0					
	2.5	7.0	2.5	11.0					
	3.0	8.3	3.0	15.0					
	3.5	10.0	3.5	21.0					
	4.0	12.0	4.0	26.0					
	4.5	14.0	4.5	31.0					
	5.0	16.0	5.0	36.0					
Butyro-lactone	0.0	0.0	0.0	0.0					
	0.5	1.0	0.5	1.0					
	1.0	2.0	1.0	4.0					
	1.5	4.0	1.5	8.0					
	2.0	5.0	2.0	15.0					
	2.5	7.0	2.5	22.0					
	3.0	9.0	3.0	28.0					
	3.5	11.0	3.5	36.0					
	4.0	13.0	4.0	44.0					
	4.5	16.0	4.5	52.0					
	5.0	18.0	5.0	60.0					

	Tetra Methyl Ammonium Chloride and Hydroxy Amine Hydrochloride		Sodium Chloride	
	E	I	E	I
Ethyl Acetate	0.0	0.0	0.0	0.0
	0.5	5.0	0.5	1.0
	1.0	10.0	1.0	3.0
	1.5	15.0	1.5	4.0
	2.0	20.0	2.0	6.0
	2.5	25.0	2.5	7.0
	3.0	30.0	3.0	9.0
	3.5	36.0	3.5	10.0
	4.0	40.0	4.0	12.5
	4.5	46.0	4.5	13.0
	5.0	52.0	5.0	14.0

TABLE III

DISCHARGE CHARACTERISTICS OF CELL TYPE Mg/KSCN/S:NH_4SCN:C/Ag

Cell	Running Time Hr.	Voltage (Average)	Current (Average) ma	Net Weight g	Coulombs	% Cathode Effec.	Watt hours lb.	Watt hours $in.^3$
Control Minus 70°F.								
(a)	17	1.53	15.35	6.12	941	10.22	29.6	1.155
(b)	11.25	1.77	17.75		720	7.84	26.2	1.02
Membrane Cation Type 4 Minus 70°F.								
(a)	31	1.54	16.7	6.20	1942	16.95	58.4	2.30
(b)	14.25	1.91	17.4		892	7.76	34.7	1.37
Membrane Cation Type 4 Room Temp.								
(a)	13	1.63	29.6	6.20	1400	12.2	40.9	1.815
(b)	7.6	1.90	34.6		870	7.57	36.7	1.445

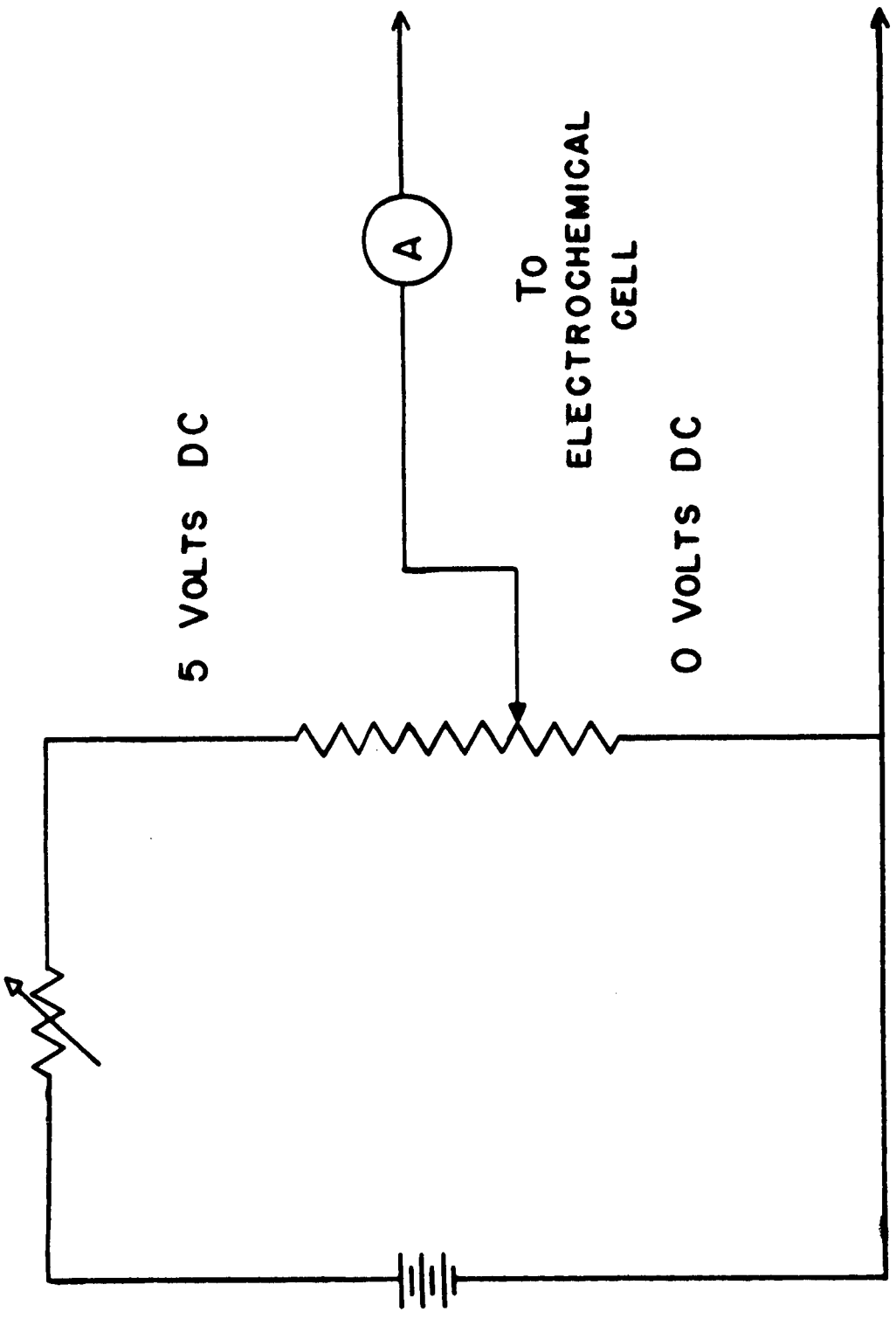
Note: (a) Cell was drained to one volt
(b) Cell was drained to 75% of peak load voltage

TABLE III (Continued)

DISCHARGE CHARACTERISTICS OF CELL Mg/KSCN/$HgSO_4$:NH_4SCN:C/Ag

Cell	Running Time Hr.	Voltage (Average)	Current (Average) ma	Net Weight g	Coulombs	% Cathode Effec.	Watt hours lb.	Watt hours in.^{3}
Control	5.63	2.26	45.5	7.12	924	53.9	37.1	1.22
Control	7.4	2.24	19.4	7.12	518	30.1	20.5	0.68
Membrane Cation Type 4	8.42	2.30	46.5	11.76	1410	80.1	34.85	2.36
Type 4	8.4	2.30	43	7.12	1300	75.6	52.7	2.16
Type 4	9.2	2.29	22	7.12	729	42.4	29.5	1.21
Membrane Cation Type 2	7.33	2.31	46	7.12	1215	70.6	49.8	2.03
Membrane Cation Type 6	8.25	2.29	43	7.12	1280	74.6	51.8	2.22
Membrane Anion Type 3	1.55	2.25	46.5	11.76	260	14.8	6.33	0.49

SCHEMATIC I



FUNCTIONAL SCHEMATIC:
DECOMPOSITION VOLTAGE MEASUREMENT

REFERENCE NO. 1 LITERATURE REVIEWED

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4 5 6 Report On: Column 3 Code Entry 5

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APPENDIX I

Deliberation on the high equivalent weight and low voltage of aqueous batteries leads to the realization that a limiting approximate relationship exists between oxidation potential and equivalent weight. In Table I of this Appendix some examples of conventional or desirable anode reactants are listed with their equivalent weights and acid aqueous electrode potentials. Figure 1 is a semi log plot of this data with the equivalent weight along the log axis. Note that lithium represents a limiting case for the alkali metal family and the points for K and Na are rejected in this derivation. Mg represents the limiting case for the alkaline earths. A line is drawn through the remaining points of Figure 1 representing mathematically the approximate anode equivalent weight as a function of anode potential.

Cathode reactants were similarly treated and the two lines combined. The equation $M = e(S - HX)$ was abstracted from the plots where M is equivalent weight, X is the electrode potential, and S and H are characterizing constants.

Appreciation is expressed to Mr. William A. Robertson of NASA for early detection and correction of an error made in the reasoning used to abstract the one equation from the two plots.

The correct values for S and H were found to be approximately 5.5 and 0.4 respectively.

More comprehensive plotting and numerical regression of the line to the points would be expected to provide additional signified figures but the above appear adequate for selection and screening of solvents.

Aqueous Oxidation Potentials vs. Standard Hydrogen Electrode.

FIGURE I

APPENDIX I

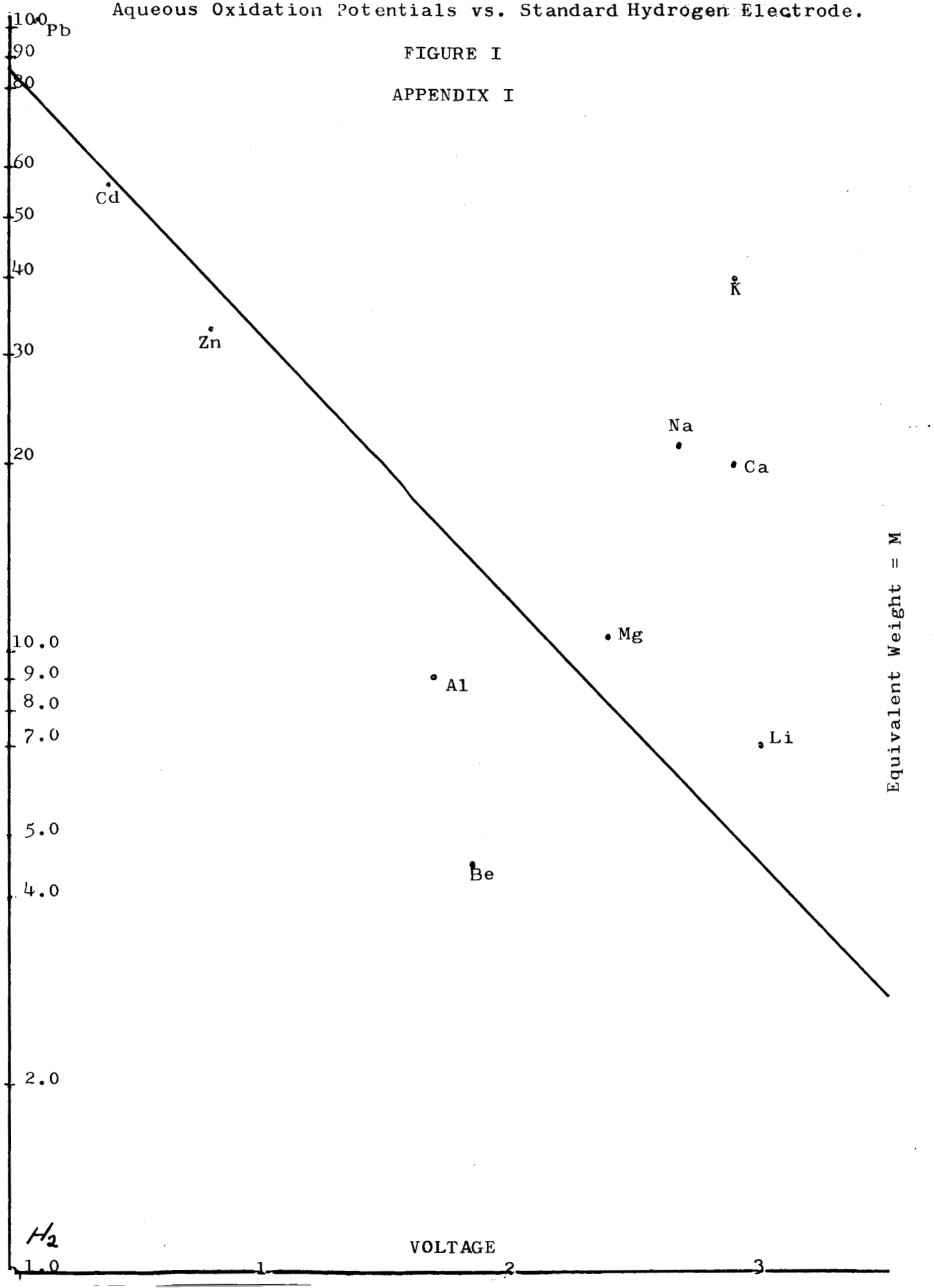


TABLE I

OF APPENDIX I

Elements	Equivalent Weight	Acid Aqueous Oxidation Potential E° (Volts)
Li	7	+3.0
Na	23	+2.7
K	39	+2.9
Mg	12	+2.4
Ca	20	+2.9
Al	9	+1.7
Be	4.5	+1.85
Zn	33	+0.8
Cd	56	+0.4
Pb	104	+0.1

APPENDIX II

L. Farkas: (2-132)

Farkas derived the following expression for the conductance of the alkali and alkaline earth metals in ammonia solution:

$$X = \frac{3 \times 10''}{J_o^{\frac{1}{2}}} \cdot 10^{-\frac{(4.2 \times 10^6 J_o^{\frac{1}{2}})}{C^{\frac{1}{3}}}}$$

where $1000\ X/c = \lambda$, the equivalent conductance, C is the concentration in moles per liter of solution, and J_o is an ionization potential which is evaluated from one experimental value of X, the conductance.

Farkas assumed that the electrons are restricted at all concentrations. Thus, in concentrated solutions, where the conductance increases rapidly, he regarded the conductance as involving nonmechanical transfer to jump from one metal atom to the next. From a theoretical standpoint, the electrons on the metal atoms required to penetrate a potential energy barrier in passing from one atom to the next and the probability that this will occur may be calculated from the quantum theory. A significant assumption in this theory is that the alkali metal atoms form an ordered structure in solution. Since an electric field is present between the electrodes (potential difference), the probability that the solvated or the free electrons will go through the potential energy barrier in the direction of the anode is greater than that for the opposite direction. Thus, a net flow of electrons through the solution results. This theory is in moderate agreement with experiments performed on solutions containing one to five moles of alkali metal ion per liter of solution.

Magnetic-susceptability measurements conducted on these solutions provide additional evidence for the rejection of the free electron in solution concept, ($Li = Li^+ + e^-$).

Assuming that the electrons are not free but are bound to alkali metal ions or to ammonia molecules, the solution would be paramagnetic and the volume susceptability would

depend on the temperature and concentration, in accordance with Curie's law:

$$K = \frac{Nu_o^2 J(J+1) g^2}{V3kT}$$

$\frac{N}{V}$ = the number of alkali metal ions or ammonia molecules NH_3 per unit volume of solution

u_o = one Bohr magneton

K = Boltzmann's constant

J_o = the resultant spin-orbital momentum quantum number of the electron

g = Lande factor = weight factor

$$= 1 + \frac{J(J+1) + S(S+1) - L(L+1)}{2J(J+1)}$$

Thus, at the alkali metal ion concentration at which the minimum is observed in liquid ammonia (0.2n), the contribution of neutral atoms would be at a maximum according to the Kraus theory. However, it is observed that in this region, the magnetic susceptability is extremely low or negative. This negativity corresponds to diamagnetism and not to paramagnetism. From the low values at medium concentrations, the magnetic susceptability per mole of alkali metal ion rises until, in highly dilute solutions, it is equivalent to a magnetic moment of one Bohr magneton per metal atom.

Qualitatively, one would expect this type of magnetic behavior if all of the electrons were free or under the influence of a periodic (in space, not time) potential field of the alkali metal atoms. At high concentrations, they would constitute a degenerate Fermi gas with a susceptability practically independent of temperature, but at low concentrations the degeneracy would be lifted, with a corresponding rise in the magnetic susceptability.

APPENDIX - III

Pauli-Block Formula Derivation

The Pauli-Block formula is derived as follows:

The Fermi-Dirac distribution law for an electron is

$$ni = \frac{1}{e^{\beta + \frac{\epsilon_1}{NkT}} + 1}$$

with $\epsilon_i = \epsilon_t + \epsilon_m$, where ϵ_t is the translational and ϵ_m the magnetic energy of an electron in a magnetic field, H. In terms of the quantum numbers n_1, n_2, n_3, m_j, J, L, S, and the weight factor (g).

We have $g = 1 + \frac{J(J+1) + S(S+1) - L(L+1)}{2J(J+1)}$

and $\epsilon_t = \frac{h^2}{8m}\left(\frac{n_1^2}{a^2} + \frac{n_2^2}{b^2} + \frac{n_3^2}{c^2}\right)$,

$abc = V$, the volume; $\epsilon_m = -m_j g u_o H$.

The magnetic moment, M, of the system of electrons will be

$$MH = -\sum_{m=-j}^{m=+j} n_m \epsilon_m = -\frac{4\pi mV}{h^3}\sqrt{2m}\,(kT)^{3/2} \sum_{m=-j}^{m=+j} \epsilon_m \int_{\pi}^{\infty} \frac{X^{\frac{1}{2}}dX}{e^{\beta + x + \epsilon_m/kT} + 1}$$

with $J = S = \frac{1}{2}$, $L = 0$, $g = 2$, $K = \frac{I}{H} = \frac{M}{HV}$, we get, after an approximate evaluation of the integrals, the Pauli-Block Relations.

$$K = u_o^2 \left[12 \left(\frac{\pi}{3}\right)^{2/3} \left(\frac{N}{V}\right)^{1/3} \frac{m}{h^2} - \left(\frac{64\pi^4}{9}\right) V \frac{m^3 k^2 T^2}{Nh^6} \right]$$

K = volume susceptability

N = Avagadro's number 6.023×10^{23} $\frac{\text{gram-atoms}}{\text{mole}}$

V = the volume available for one mole of electrons

m = mass of the electron

h = Planck's constant = 6.6238×10^{-27} erg-second

k = Boltzmann's constant = $\frac{R}{N} = 1.380 \times 10^{-16}$ erg. per degree C.

T = Absolute temperature

The second term on the right of the Pauli-Block formula containing T^2 is usually very small. For an electron gas,

$$K = 2.20 \times 10^{-14} \left(\frac{N}{V}\right)^{1/3} - 1.03 \times 10^{7} \left(\frac{V}{N}\right) T^2 ;$$

and since $\frac{N}{V} = 2.6 \times 10^{22}$ for metallic sodium,

$$K = 6.6 \times 10^{-7} - 3.9 \times 10^{-16} T^2.$$

K is practically independent of temperature. For most metals and solutions of metals in liquid ammonia, $\frac{N}{V} = 10^{22}$ to 10^{16}; only in the case of 10^{16} does the second term become important. (K) is the volume susceptibility; X_m, (the molal susceptibility) is the molar volume times (K). Substituting in the low temperature expression for (K) and omitting the small second term, we get,

$X_m = 1.86 \times 10^{-6} \; V^{2/3}$, where V is the volume available to one mole of electrons. This is the paramagnetic susceptibility.

For electrons in a periodic field:

At low temperature

$$K = K_2 \quad u_o^2 \frac{N}{V\beta}$$

$K_2 \cong 1$; β = exchange energy of the electron going from one potential hole to a neighboring one.

Electrons in a periodic field at high temperature

$$K = N \frac{u_o^2}{VkT}$$

FIGURE I

Voltage versus Time for
Ammonia Battery at Minus 70°F.
For the System
$Mg/KSCN/HgSO_4 \cdot 2NH_3 + NH_4CNS + C/Ag$

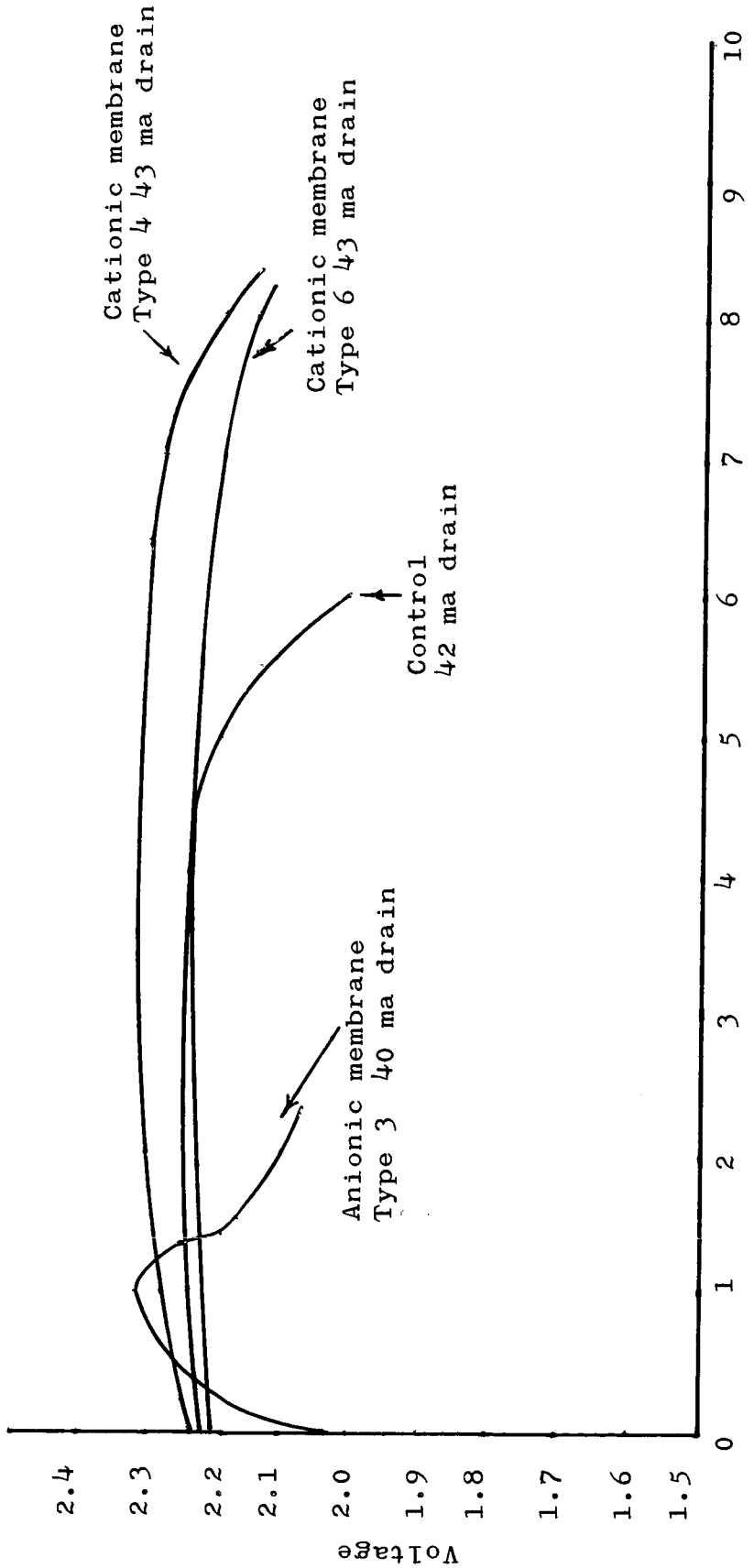
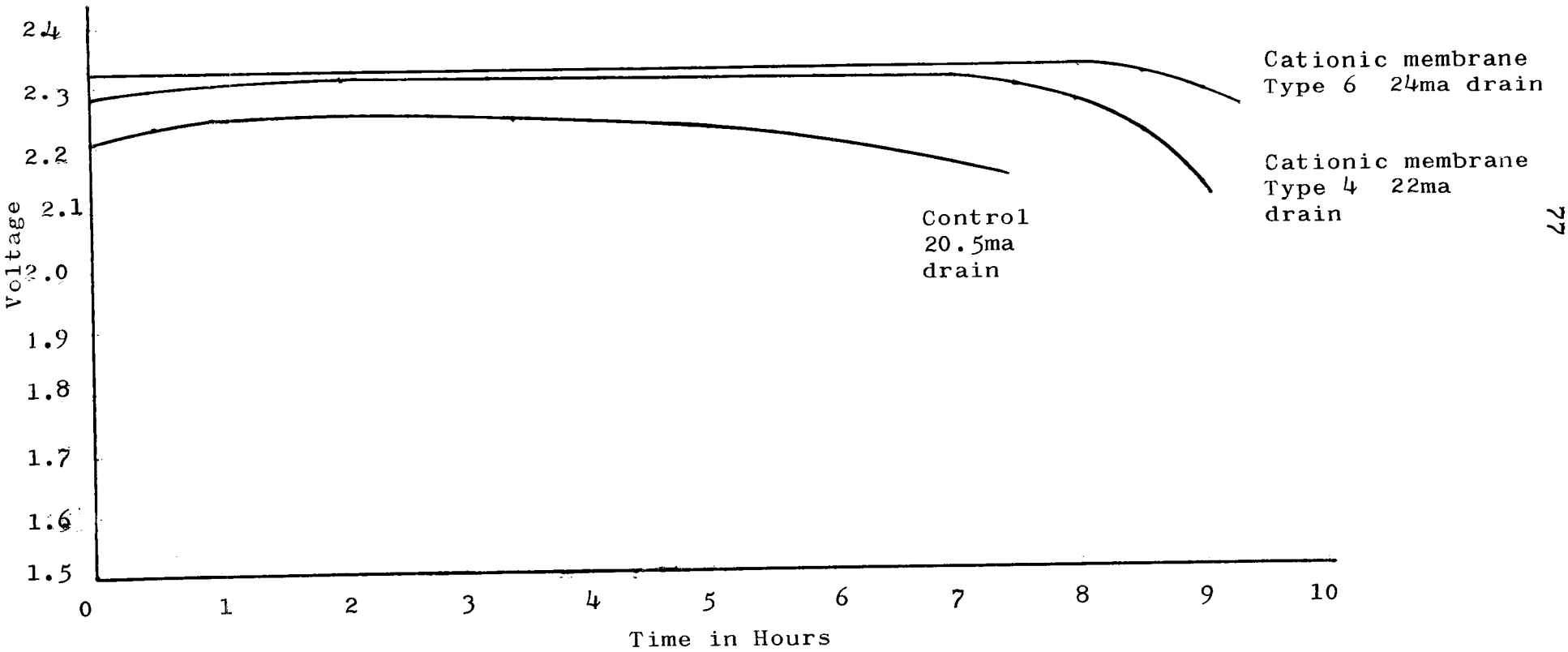


FIGURE II

Voltage versus Time for
Ammonia Battery at
minus 70°F for the
System Mg / KSCN / $HgSO_4 \cdot 2NH_3$ + NH_4CNS + C / Ag



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